Migration of Contaminated Soil and Airborne Particulates to Indoor Dust

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Received March 3, 2009. Revised manuscript received August 31, 2009. Accepted September 3, 2009.

We have developed a modeling and measurement framework for assessing transport of contaminated soils and airborne particulates into a residence, their subsequent distribution indoors via resuspension and deposition processes, and removal by cleaning and building exhalation of suspended particles. The model explicitly accounts for the formation of house dust as a mixture of organic matter (OM) such as shed skin cells and organic fibers, soil tracked-in on footwear, and particulate matter (PM) derived from the infiltration of outdoor air. We derived formulas for use with measurements of inorganic contaminants, crustal tracers, OM, and PM to quantify selected transport parameters. Application of the model to residences in the U.S. Midwest indicates that As in ambient air can account for nearly 60% of the As input to floor dust, with soil track-in representing the remainder. Historic data on Pb contamination in Sacramento, CA, were used to reconstruct sources of Pb in indoor dust, showing that airborne Pb was likely the dominant source in the early 1980s. However, as airborne Pb levels declined due to the phase-out of leaded gasoline, soil resuspension and track-in eventually became the primary sources of Pb in house dust.

Introduction

Inorganic contaminants can migrate to indoor environments via infiltration of outdoor air containing suspended PM and track-in of soil adhering to footwear (*1, 2*). Examples include the transfer of contaminants into houses derived from atmospheric emissions (*3*) and metals associated with mining operations (*4*). After entering a residence, inorganic substances present in soil and airborne particles become incorporated into household dusts, which serve as a primary reservoir for such substances (*5*). Young children can have elevated contaminant exposures because they exhibit behaviors that increase indirect ingestion by way of hand-to-mouth activities and mouthing of various dust-contaminated objects, and ingest dust at rates that are greater than adults on a body-weight basis (*6*). For contaminants such as As and Pb indirect ingestion often constitutes the principal exposure mechanism for children (*4, 7*), and therefore health-risk assessments need to address transport mechanisms that bring contaminants indoors as well as the factors that influence contaminant concentrations on contact surfaces indoors. Of special interest are fractionation/dilution processes applicable to soil/dust particles that adhere to hands and footwear and the implications for soil/dust sampling. It is also important to characterize relationships between contaminant source terms and outdoor-to-indoor transport processes to design, implement, and evaluate measures for managing health risks at contaminated sites (e.g., Superfund sites).

Conceptual frameworks (*3, 8*) and models for simulating transport processes involving contaminated soils and indoor dusts have been presented by several investigators (*1, 8*–*10*). These studies, however, were not specifically designed to support assessments of dermal contacts with house dusts containing inorganic substances derived from both outdoor air and exterior soils. We have therefore devised a modeling and measurement framework to address the primary exposure media in residences related to indirect ingestion, consisting of floor dust and dust fall landing on horizontal contact surfaces. Because of the key role of house dust as both a transport and exposure medium, we simulate its formation as well as its redistribution indoors via resuspension and deposition processes and removal by cleaning. To guide the process of model parametrization, we present equations that are used in conjunction with site-specific measurements of inorganic substances in indoor and outdoor media to quantify model parameters involving particle transport and building properties. We also conduct sensitivity/uncertainty analyses to identify which parameters have the greatest impact on model predictions.

Methods

Movement of inorganic substances to the indoor environment and subsequent incorporation and redistribution in indoor dusts, as depicted in Figure 1, is governed largely by human activities, the built environment, and the nature and sources of outdoor contamination. Track-in of soil on footwear, for example, is a transport pathway that is a function of the number of adults in a household, amount of time children spend outdoors, etc. (*11*). In addition, walking and other activities resuspend floor/carpet particles to indoor air, which are then redeposited on indoor surfaces and vented from the house via air exchange processes (*1*). Cleaning activities remove dust accumulations on indoor surfaces (and also resuspend/redistribute dust), but their frequency and effectiveness varies according to cleaning devices used, types of floor surfaces (*12*), and cleaning behaviors of residents. As the duration between cleaning activities increases, floor dust loading (i.e., mass of dust per unit surface area) increases, as does resuspension of floor particles because of increased mass of resuspendable floor dust.

Relevant characteristics of the built environment affecting the flux of suspended outdoor particles to indoor air include the air exchange rate of a house and effectiveness of the building shell in filtering out airborne particles. Filters in air handling units will also remove suspended PM. Indoor air volume (determined by ceiling height and floor area) controls in part the predicted concentration of airborne particles. Floor area influences the magnitude of dust loading and retention on floors. Particles penetrating the building shell are subsequently deposited onto horizontal surfaces or vented from the house. The chemical composition of both suspended dust and dust fall is a function of particles that have penetrated the building shell and particles resuspended from floor surfaces—which are a mix of particles from tracked-in soil, indoor sources of OM, and deposited particles derived from outdoor air.

Transport Relationships. The transfer of inorganic substances in outdoor air and soil to the interior of a residence,

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10.1021/es9003735 CCC: $40.75
Published on Web 09/24/2009

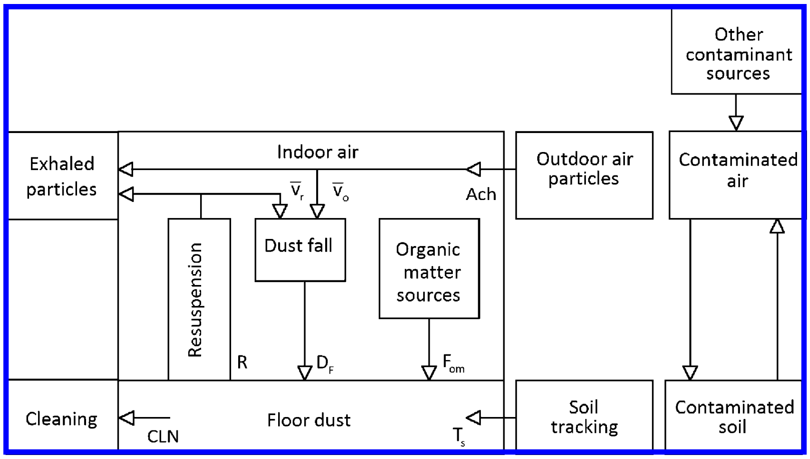

FIGURE 1. Conceptual diagram depicting the movement of contaminated soil and airborne particulates into a residence, subsequent mixing with organic matter in floor dust, redistribution indoors via resuspension, and removal by cleaning and exhalation with building air. The variables are defined as follows: CLN is the first-order particle removal rate from floors due to cleaning activities, R is the resuspension rate of floor particles to indoor air, D_F is the rate of dust fall to floors, F_{om} is the flux of organic matter onto floor surfaces, T_s is the rate of soil track-in on footwear into a residence, Ach is the air exchange rate, and $\bar{v}_r$ and $\bar{v}_o$ are the mean deposition velocities for resuspended particles and outdoor-derived air particles, respectively.

and their subsequent fate in indoor dust can be formulated as a two-compartment model consisting of an air compartment linked to a floor compartment via resuspension and deposition processes. We have defined a system of time-varying mass-balance relationships that simulate dust fall and the bulk accumulation and loss of particles on floor surfaces together with the associated concentration of an inorganic contaminant/tracer in those exposure media (See Supporting Information for derivation). PM entering the indoor environment is assumed to be uniformly mixed in the indoor air of a naturally ventilated residence and evenly distributed on floor surfaces. Emissions of submicrometer particles from smoking and cooking represent minor contributors to dust due to their low deposition loss rates (*13*), and therefore they are not addressed in the model. We define the flux of OM to floors as a lumped parameter to represent the input of lint, skin particles, organic fibers, food debris, etc. to floors. The concentration (g g^{-1}) of OM in dust as well as soil and airborne PM is operationally defined by the mass loss on ignition (*14*).

As a means of examining the transport processes addressed in the mass-balance relationships, we have derived steady-state solutions for selected model parameters. The mass loading of dust on floor surfaces (M_{fl} in g m^{-2}) under steady-state conditions is a function of (a) inputs to floor surfaces from the track-in of soils on footwear, OM fluxes from indoor sources, and the deposition of suspended dust, and (b) removal by cleaning and resuspension, or

$$M_{fl} = \frac{T_s + F_{OM} + D_F \times A_{fl}}{A_{fl}(R + CLN)} \qquad (1)$$

where T_s is the rate of soil track-in on footwear to a residence (g d^{-1}); F_{om} is the flux of OM onto floor surfaces (g d^{-1}); D_F is the rate of dust fall to floors (g m^{-2} d^{-1}); A_{fl} is the floor area of a residence (m^2); R is the resuspension rate of floor particles to indoor air (d^{-1}); and CLN defines the first-order particle removal rate from floors due to cleaning activities (d^{-1}). The equation for determining the rate of dust fall indoors is given by

$$D_F = \frac{P \cdot Ach \cdot H \cdot TSP_o}{(Ach \cdot H + \bar{v}_o)}\bar{v}_o + \frac{R \cdot M_{fl}}{(Ach \cdot H + \bar{v}_r)}\bar{v}_r \qquad (2)$$

The first term represents the deposition of outdoor-derived PM that penetrates the building shell, where Ach is the air exchange rate (d^{-1}) for a residence; TSP_o is total suspended PM in outdoor air (g m^{-3}); H is the ceiling height (m); P is the dimensionless penetration factor representing the particle removal efficiency of the building shell (where $0 \le P \le 1$); and $\bar{v}_o$ is the mean deposition velocity (m d^{-1}) of outdoor-derived PM settling onto floors. The second term in eq 2 quantifies the deposition rate of airborne dust generated by the resuspension of floor dust by human activities (i.e., walking, cleaning, vacuuming, etc.), where $\bar{v}_r$ is the mean deposition velocity (m d^{-1}) of resuspended dust settling on floors. The dust fall rate can also be calculated as the product of the concentration of TSP in indoor air (denoted TSP_{in} in g m^{-3}) and the mean deposition velocity of suspended dust particles (denoted $\bar{v}_{in}$ in m d^{-1}), that is, $D_F = TSP_{in} \times \bar{v}_{in}$.

The concentration of an outdoor-derived inorganic substance in dust fall, denoted C^s_{df} (μg g^{-1}), is given by

$$C^s_{df} = \frac{Ach \cdot H \cdot C^s_{tsp\_o} \cdot P \cdot TSP_o \cdot \bar{v}_o(Ach \cdot H + \bar{v}_r) + C^s_{fl} \cdot M_{fl} \cdot R \cdot \bar{v}_r(Ach \cdot H + \bar{v}_o)}{Ach \cdot H \cdot P \cdot TSP_o \cdot \bar{v}_o(Ach \cdot H + \bar{v}_r) + M_{fl} \cdot R \cdot \bar{v}_r(Ach \cdot H + \bar{v}_o)} \qquad (3)$$

where $C^s_{tsp\_o}$ is the bulk concentration of the substance in suspended outdoor air particles (μg g^{-1}) and C^s_{fl} is its concentration in floor dust (μg g^{-1}), which can be calculated from

$$C^s_{fl} = \frac{\bar{v}_o(C^s_{om} \cdot F_{om} + C^s_s \cdot T_s) + Ach \cdot H(C^s_s \cdot T_s + A_{fl} \cdot P \cdot C^s_{tsp\_o} \cdot TSP_o \cdot \bar{v}_o)}{\bar{v}_o(F_{om} + T_s + Ach \cdot A_{fl} \cdot P \cdot TSP_o \cdot H) + Ach \cdot H(F_{om} + T_s)} \qquad (4)$$

where C^s_s is the concentration of the substance in outdoor soil (μg g^{-1}) and C^s_{om} is its concentration in OM (μg g^{-1}). An alternative equation for determining M_{fl} that does not use D_F as an input parameter is

$$M_{fl} = \frac{(\bar{v}_r + Ach \cdot H)(\bar{v}_o(F_{om} + T_s) + Ach \cdot H(F_{om} + T_s + A_{fl} \cdot P \cdot TSP_o \cdot \bar{v}_o))}{A_{fl}(\bar{v}_o + Ach \cdot H)(CLN(\bar{v}_r + Ach \cdot H) + R \cdot Ach \cdot H)} \qquad (5)$$

The product of C^s_{fl} and M_{fl} represents the loading of a substance on floor surfaces, denoted W^s_{fl} (μg m^{-2}).

Contaminant inputs/outputs for floors (in μg d^{-1}) via deposition from outdoor-derived air PM (denoted IN_{air}) plus soil tracking (IN_{track}) and subsequent removal by building exhalation of resuspended particles (OUT_{exh}) and cleaning (OUT_{cln}) are calculated from these formulas:

$$IN_{air} = \frac{Ach \cdot P \cdot H \cdot C^s_{air}}{(\bar{v}_o + Ach \cdot H)} \bar{v}_o \cdot A_{fl} \tag{6}$$

$$IN_{track} = C^s_s \cdot T_s \tag{7}$$

$$OUT_{exh} = \frac{M_{fl} \cdot C^s_{fl} \cdot R}{(\bar{v}_r + Ach \cdot H)} Ach \cdot H \cdot A_{fl} \tag{8}$$

$$OUT_{cln} = CLN \cdot A_{fl} \cdot M_{fl} \cdot C^s_{fl} \tag{9}$$

where C^s_{air} is the contaminant concentration in air (μg m^{-3}) and equals $C^s_{tsp\_o} \times TSP_o$.

Parameter Estimation. Several parameters in our analytical formulation cannot be measured directly or are otherwise difficult to quantify. We have therefore developed a series of formulas that can be used in conjunction with site-specific measurements of crustal soil tracers or inorganic contaminants in indoor and outdoor media along with TSP levels in indoor and outdoor air, dust loading on floors, rate of dust fall, and ceiling height. If the air exchange rate for residences can be specified, then the penetration factor is calculated as

$$P = \frac{D_F(C^s_{df} - C^s_{fl}) - Ach \cdot H \cdot TSP_{in}(C^s_{fl} - C^s_{tsp\_in})}{Ach \cdot H \cdot TSP_o(C^s_{tsp\_o} - C^s_{fl})} \tag{10}$$

Otherwise, if P can be specified, then

$$Ach = \frac{D_F(C^s_{df} - C^s_{fl})}{TSP_{in} \cdot H(C^s_{fl} - C^s_{tsp\_in}) + P \cdot H \cdot TSP_o(C^s_{tsp\_o} - C^s_{fl})} \tag{11}$$

The resuspension rate, given P calculated from eq 10, is

$$R = \frac{D_F + Ach \cdot H(TSP_{in} - P \cdot TSP_o)}{M_{fl}} \tag{12}$$

Deposition velocity estimates for outdoor-derived particles and resuspended particles indoors are

$$\bar{v}_o = -\frac{D_F(C^s_{df} - C^s_{fl})}{TSP_{in}(C^s_{fl} - C^s_{tsp\_in})} \tag{13}$$

$$\bar{v}_r = \frac{D_F(C^s_{tsp\_o} - C^s_{df})}{TSP_{in}(C^s_{tsp\_o} - C^s_{tsp\_in})} \tag{14}$$

Dual measurements of the concentrations of OM and inorganic substances (i.e., crustal tracers and/or contaminants) in indoor and outdoor media together with data on building properties, TSP_o, and $\bar{v}_o$ can be used to determine the OM flux to floors and the soil track-in rate:

$$F_{om} = \frac{Ach \cdot A_{fl} \cdot H \cdot P \cdot TSP_o \cdot \bar{v}_o(C^s_s(C^{om}_{fl} - C^{om}_{tsp\_o}) + C^s_{tsp\_o}(C^{om}_s - C^{om}_{fl}) + C^s_{fl}(C^{om}_{tsp\_o} - C^{om}_s))}{(\bar{v}_o + Ach \cdot H)(C^s_s(1 - C^{om}_{fl}) + C^s_{fl}(C^{om}_s - 1) + C^s_{om}(C^{om}_{fl} - C^{om}_s))} \tag{15}$$

$$T_s = \frac{Ach \cdot A_{fl} \cdot H \cdot P \cdot TSP_o \bar{v}_o(C^s_{tsp\_o}(1 - C^{om}_{fl}) + C^s_{fl}(C^{om}_{tsp\_o} - 1) + C^s_{om}(C^{om}_{fl} - C^{om}_{tsp\_o}))}{(\bar{v}_o + Ach \cdot H)(C^s_{fl}(1 - C^{om}_s) + C^s_s(C^{om}_{fl} - 1) + C^s_{om}(C^{om}_s - C^{om}_{fl}))} \tag{16}$$

where C^{om}_{fl}, C^{om}_s, and $C^{om}_{tsp\_o}$ are the concentrations of OM in floor dust, soil, and outdoor TSP (g g^{-1}).

Soil Resuspension Model. Contaminants deposited onto surficial, undisturbed soils are subject to redistribution via aeolian resuspension (*15*), which constitutes a potential source term for the indoor environment due to infiltration of suspended particles across the exterior shell of a residence. In general, resuspension decreases with time as the deposited contaminant undergoes weathering processes (e.g., incorporation of the contaminant within a soil matrix, vertical migration in soil, etc.) that reduces the erodability of a surficial soil contaminant. The concentration of a soil contaminant in air due to resuspension can be determined from (*16*)

$$C^s_{air} = C^s_L \cdot S_f \tag{17}$$

where C^s_{air} is the concentration of a soil contaminant in outdoor air (μg m^{-3}), C^s_L is the accumulated deposition of a contaminant per unit surface area of soil (μg m^{-2}), and S_f is the resuspension factor (m^{-1}). The value of C^s_L can be calculated as the product of C^s_s, the soil depth applicable to resuspension, d (m), and soil density, ρ (g m^{-3}). For aged soil sources the value of S_f is approximately 10^{-9} m^{-1} (*16*).

Model Assessment. As a means of analyzing the factors controlling transfer of soil and airborne contaminants to residential environments, we used the assessment framework to reconstruct transport mechanisms associated with residential locations in the U.S. and The Netherlands. Information on the U.S. residences is from the National Human Exposure Assessment Survey (NHEXAS) involving six Midwestern states (*17*) and a study of Pb contamination involving residences in Sacramento, CA (*18*). Residences in The Netherlands were from a neighborhood near a secondary Pb smelter in the city of Arnhem (*19*). A detailed description of the multimedia sampling performed for each study is provided in the Supporting Information.

Results and Discussion

Transport Parameters. We used monitoring data collected for the NHEXAS Midwest and Arnhem residences to estimate both building (i.e., penetration factor and air exchange rate) and transport parameters (i.e., resuspension rate and particle deposition velocities) utilizing eqs 10−14. The NHEXAS data set included information on the smoking status of households, which we used to identify nonsmoking households that had data on the concentrations of As in both indoor dust and airborne TSP. Analyses of the relevant concentration data indicated that they can be represented by log-normal probability distributions (see Supporting Information). The geometric mean (GM) concentrations of TSP in indoor and outdoor air were 2.8×10^{-5} and 2.4×10^{-5} g m^{-3}, respectively, while the associated As concentrations in the suspended PM ($C^s_{tsp\_in}$ and $C^s_{tsp\_o}$) were 15 μg g^{-1} and 27 μg g^{-1}. The geometric mean rate of dust fall (D_F) was 3.0×10^{-3} g m^{-2} d^{-1} and the related As content of dust fall (C^s_{df}) was 7.4 μg g^{-1}. The dust loading on floors (M_{fl}) was 0.28 g m^{-2}. Clayton et al. (*17*) estimated a median concentration of As in the floor dust of NHEXAS residences (C^s_{fl}) as 5.8 μg g^{-1}. For comparison, the GM concentration of As in soils (C^s_s) in the eastern U.S. is 4.8 μg g^{-1} (*20*)—a factor of 5.6 lower than its concentration in airborne particles. The elevated value of $C^s_{tsp\_o}$ likely reflects the impact of atmospheric As emissions from fossil fuel combustion (*21*).

Assuming a ceiling height of 2.4 m (default value for all analyses) and an air exchange rate equal to 8.6 d^{-1} for houses in the climate zones associated with Midwestern states (*22*), the estimated value of P is 0.96 (from eq 10). The deposition velocity for outdoor-derived particles infiltrating the residences ($\bar{v}_o$) is equal to 18.6 m d^{-1} (eq 13), whereas the deposition velocity predicted for resuspended particles ($\bar{v}_r$) is 175 m d^{-1} (eq 14). The deposition velocity for TSP_{in} ($\bar{v}_{in}$) is 107 m d^{-1} and the estimated resuspension rate (R) is 0.011 d^{-1} (eq 12). The value for P is near its upper limit of 1, which means that the ambient As-bearing aerosols were able to effectively penetrate the building shells of the residences surveyed. The estimated value of $\bar{v}_o$ for outdoor-derived particles falls within the range of 14−26 m d^{-1} reported in Thatcher and Layton (*1*) for particles 1−5 μm in diameter, which corresponds to the sizes of most crustal elements in airborne particulates (*23*). Our estimate of $\bar{v}_r$ is also within the range of 135 and 234 m d^{-1} they reported for the larger particles associated with resuspension (i.e., 10−25 μm and >25 μm in diameter, respectively). We also note that our reconstructed value for R is a daily value, and thus resuspension rates for active, nonresting hours of residents will be higher (*24*).

The Arnhem study provided data on the Pb composition of indoor and outdoor TSP as well as indoor dusts for about 100 houses in a single neighborhood. Study results were undifferentiated by smoking status of the households, and the ratio of TSP_{in} (1.20×10^{-4} g m^{-3}) to TSP_o (6.40×10^{-5} g m^{-3}) was 1.88, compared with a ratio of about 1.17 for the smoke-free Midwestern residences. To compensate for smoking emissions, we divided the reported dust fall rate of 7.7×10^{-3} g m^{-2} d^{-1} (which is dominated by resuspended dust particles) by 107 m d^{-1} (deposition velocity of TSP_{in} for the Midwest housing) to obtain an adjusted TSP_{in} value of 7.2×10^{-5} g m^{-3}. We also increased the value of $C^{s}_{tsp\_in}$ from 2.29×10^3 to 3.6×10^3 μg g^{-1} to account for the decreased mass of suspended PM in indoor air. The reported GM loading of dust on floors was 0.255 g m^{-2}, while the concentrations of Pb in floor dust, dust fall, and TSP_o were 482, 1.00×10^3, and 6.4×10^3 μg g^{-1}, respectively (see Supporting Information).

The reconstructed Ach value for the Arnhem residences of 10.8 d^{-1} (from eq 11, with P = 1 to represent older, nonenergy efficient housing) falls within the 10th and 50th percentile Ach values (7 and 14 d^{-1}) reported for a sample of Dutch homes (*25*). The computed indoor deposition velocities (17.8 and 206 m d^{-1} for $\bar{v}_o$ and $\bar{v}_r$) are comparable to the results for the NHEXAS residences, but the estimated resuspension rate of 0.031 d^{-1} is considerably higher. One explanation for this difference is the vacuum cleaning that occurred "once every 1 or 2 days" in the Arnhem residences (*19*). Vacuuming produces a significant increase in airborne PM that is greater than 10 μm in diameter (*26, 27*), and the elevated value of $\bar{v}_r$ for the Arnhem residences may reflect this fractionation process. Differences in the types of flooring between the Arnhem and NHEXAS Midwest residences further complicate comparisons between the resuspension characteristics of the housing. In addition, the mass loading data for the NHEXAS residences were based on wipe samples of flat floor surfaces, but resuspended dust from carpeted surfaces undoubtedly contributed to indoor TSP levels as well. Carpets retain more dust than bare floors (*28*) and resuspension rates for carpeting are also greater than those for bare floors (*24*).

Estimates of OM Fluxes and Soil Track-In. Indoor dust is a mixture of soil tracked into a residence, PM derived from ambient outdoor air, and importantly, OM inputs from various sources. A prominent feature of indoor dust is its OM content, with levels of about 40 wt % in residential housing (*3, 29*). We estimated the values of F_{om} and T_s for the Midwest residences using eqs 15 and 16 based on a reference OM content of floor dust (C^{om}_{fl}) of 0.4 g g^{-1}; A_{fl} equal to 110 m^2; OM contents of TSP_o ($C^{om}_{tsp\_o}$) and soils (C^{om}_{s}) equal to 0.13 and 0.02 g g^{-1}; respectively (see Supporting Information); C^{s}_{om} equal to zero; and other input parameters as previously defined. The resulting estimates of F_{om} and T_s are 0.074 and 0.099 g d^{-1}, respectively. The amount of soil tracked into residences, as measured by accumulations on entry way mats, is a complex function of housing occupancy, weather and soil conditions, etc. (*11, 30*). Our analysis of soil track-in data (see Supporting Information) indicates that a log-normal distribution with a geometric mean value of 0.1 g d^{-1} (geometric standard deviation (GSD) = 3) can be used to characterize soil-to-house transfers via foot traffic. The reconstructed track-in value of 0.099 g d^{-1} for the Midwest residences compares favorably with the value based on soil tracking measurements. Less is known about the composition of OM in house dust on a weight basis, however, skin particles and organic fibers appear to be major constituents. For example, skin cells are constantly shed from people (*31*) and they are high in N (*32*), which suggests that exfoliated skin is a potential source of the elevated N content of house dust (*33*). Scanning electron microscopy of house dust samples has shown that dust contains many fibers ranging from less than 10 to over 100 μm in size, which are largely destroyed using high-temperature oxidation—indicating that they are predominantly organic in composition (*34*).

Contaminant Inputs and Removal. Accumulation of contaminants on floors and other surfaces depends on the magnitude of contaminant inputs to a residence and subsequent removal by cleaning and exhalation of resuspended particles from the building. We used the input−output relationships defined in eqs 6−9 along with the As media concentrations, and the transport and housing parameters developed for the Midwest residences to determine As inputs to and outputs from their floor surfaces. Soil track-in accounted for 0.48 μg d^{-1} of As input to floors, while floor deposition of As in outdoor PM derived from infiltration amounted to 0.67 μg d^{-1} (58% of total). The primary parameter controlling removal of floor dust is the cleaning loss rate (CLN), equal to 0.0053 d^{-1} (derived from eq 1). The associated residence time of floor particles ($\tau = (R + CLN)^{-1}$) is 61 d, which is less than an 85 d residence time based on a simulated weekly cleaning scenario presented in Qian et al. (*10*), but greater than a 29 (±1) d residence time given in Allott et al. (*3*) for a house in the U.K. where vacuum cleaning occurred on almost a daily basis. Significantly, over 80% of the As-bearing floor dust was removed by cleaning. Although particle resuspension and then building exhalation to outdoor air constitutes a relatively minor removal pathway for floor particles, resuspension−deposition serves as the primary redistribution mechanism of floor dust and contaminants in the indoor environment—accounting for more than 90% of the deposition flux of particles. Meyer et al. (*35*) found that dust loading rates in a sample of German residences were directly related to the number of occupants, supporting the linkage between resuspension and deposition processes.

Sutton et al. (*18*) conducted a study of Pb contamination at residences in Los Angeles, Oakland, and Sacramento, CA, from 1987 to 1991 and found that Pb in paint was a poor predictor of Pb in indoor dust. To evaluate the potential magnitude of Pb in outdoor air as an alternative source of Pb in the Sacramento residences, we reconstructed the inputs of airborne Pb to floors (IN_{air}) and inputs of Pb from soil track-in (IN_{track}) for the years 1982 and 1992. These years encompass a period during which Pb emissions to the atmosphere decreased significantly due to the phase-out of leaded gasoline. Parameter values for Ach, P, A_{fl}, and $\bar{v}_o$ used

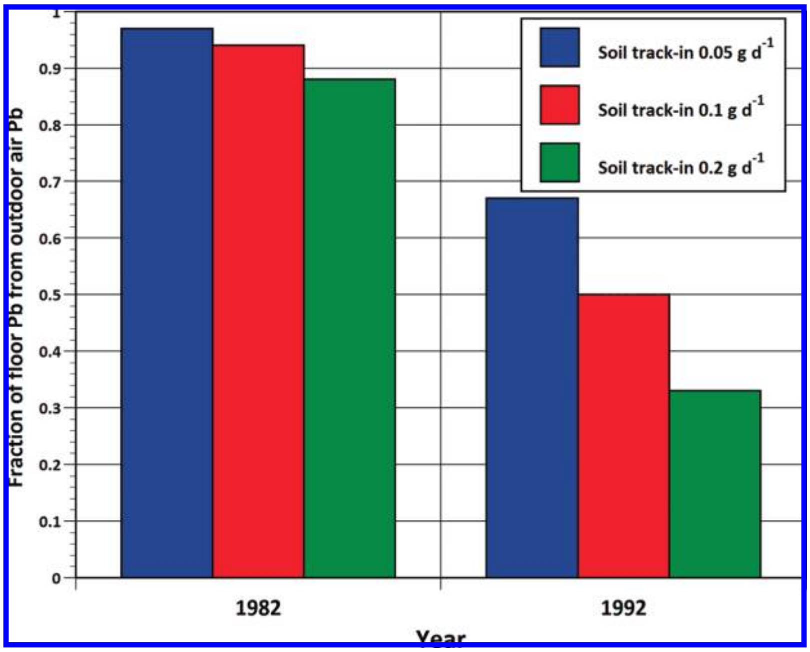

FIGURE 2. Reconstruction of the contributions of outdoor air Pb to total Pb in floor dust for three scenarios of soil track-in to Sacramento residences.

to compute IN_{air} (eq 6) are 11 d^{-1}, 1; 110 m^2, and 18 m d^{-1}, respectively, while the values of C^s_{air} for 1982 and 1992 are 0.30 and 0.020 μg m^{-3} (see Supporting Information). The resulting estimates for IN_{air} dropped from 350 μg d^{-1} in 1982 to 24 g d^{-1} in 1992. To bracket the range of soil track-in rates, we used rates of 0.05, 0.1, and 0.2 g d^{-1}. With C^s_s equal to 234 μg g^{-1} (*18*), the alternative IN_{track} values are 12, 23, and 47 μg d^{-1} of Pb. Figure 2 plots the fractions of total Pb input to floors attributable to Pb derived from outdoor air (i.e., $IN_{air} \div (IN_{track} + IN_{air})$). Airborne inputs of Pb to floors in 1982 were likely derived from both direct automotive emissions as well as secondary emissions from soil resuspension (*36*). By 1992, though, soil resuspension alone could account for airborne Pb because the associated resuspension factor of 1×10^{-9} m^{-1} calculated from eq 17 (with C^s_{air} equal to 0.020 μg m^{-3} and C^s_L equal to 1.9×10^7 μg m^{-2} based on $C^s_s = 234$ μg g^{-1} (*18*), $d = 0.05$ m, and $\rho = 1.6 \times 10^6$ g m^{-3} (*37*)) is consistent with an aged soil source (*16*).

Uncertainty/Sensitivity Analyses. Variation in the concentrations of an inorganic substance in house dust depends largely on the nature and magnitude of its sources together with modifying factors involving occupant characteristics as well as dust and building properties. To illustrate, lead loading on floors is a key determinant of blood-lead levels in children (*7*) and because contaminant loading (W^s_{fl}) is the product of M_{fl} and C^s_{fl}, the variability in W^s_{fl} is a function of the variances in the two input parameters. Assuming that C^s_{fl} and M_{fl} are statistically independent and log-normally distributed, the variance in W^s_{fl} is equal to $(\ln \mathrm{GSD}\ W^s_{fl})^2 = (\ln \mathrm{GSD}\ C^s_{fl})^2 + (\ln \mathrm{GSD}\ M_{fl})^2$. The GSDs of W^s_{fl} and C^s_{fl} for Pb in the dusts of the Sacramento residences were 4.4 and 2.3, respectively (*18*), and thus the GSD of M_{fl} is calculated as 3.4. Based on these GSD values, 68% of the variance in W^s_{fl} is accounted for by the variability in M_{fl} and the remaining 32% is associated with C^s_{fl}. This apportioning of variance is consistent with the dominant role of human factors in controlling M_{fl} via soil track-in, OM fluxes, resuspension, and cleaning activities.

To analyze the influence of model parameters on W^s_{fl} as well as the As content in dust fall, we conducted sensitivity analyses of the changes in these parameters using data for the Midwest residences. The analyses are based on low-to-high deviations from base-case values for the relevant input parameters. For all of the model parameters except P, we used the 10th and 90th percentiles of the parameter-specific log-normal distributions to represent the low and high values, while the GM defined the base-case values (see Supporting Information for parameter values). We assigned a P value of 0.95 for the base case and 1 for its upper limit, and used a value of 0.9 to define the lower limit. As shown in Figure 3, the cleaning rate is the most important parameter controlling W^s_{fl} (calculated using eq 4 for C^s_{fl} and eq 5 for M_{fl}) due in part to our specification of a broad spectrum of cleaning rates. Soil track-in is another human-related parameter that exerts a strong influence on As accumulation on floors. Additional parameters controlling W^s_{fl} listed in order of importance are the concentration of As in soil, the resuspension rate, As in outdoor TSP, and floor area. Interestingly, W^s_{fl} is insensitive to changes in the flux of OM to floors because it simultaneously reduces the As concentration in floor dust (by dilution) and increases dust loading. In contrast, the As content of dust fall (from eq 3) is most sensitive to changes in the measured As content of floor dust (and indirectly its controlling parameters such as soil track-in, OM loading, etc.), resuspension-related parameters (i.e., R and M_{fl}), and parameters involving the airborne transfer of As to the indoor environment (i.e., $C^s_{tsp\_o}$, Ach, and TSP_o).

Parameter Characterization. An important consideration regarding soil track-in is the fractionation caused by differential particle adherence to and deposition from footwear because it can produce a significant misalignment between the concentration of a contaminant measured in outdoor soil and in the soil actually tracked indoors (*38, 39*). Dust adhering to soles of the footwear of people entering a museum, for example, exhibited a bimodal distribution with peaks at 28 and 64 μm, while the greatest depletion of particles (measured on exiting the museum) was in the 50−64 μm in size range, with smaller losses extending to about 150 μm (*30*). The mass transfer of a soil contaminant on footwear will thus depend on the chemical content of such "trackable" particles, which is controlled in part by the source(s) of soil contamination.

Information on soil and dust transport processes as well as data on particle adherence to hands supports the use of two basic particle size classes to characterize soils and dusts: a fine fraction consisting of particles $\leq$60 μm in size and a

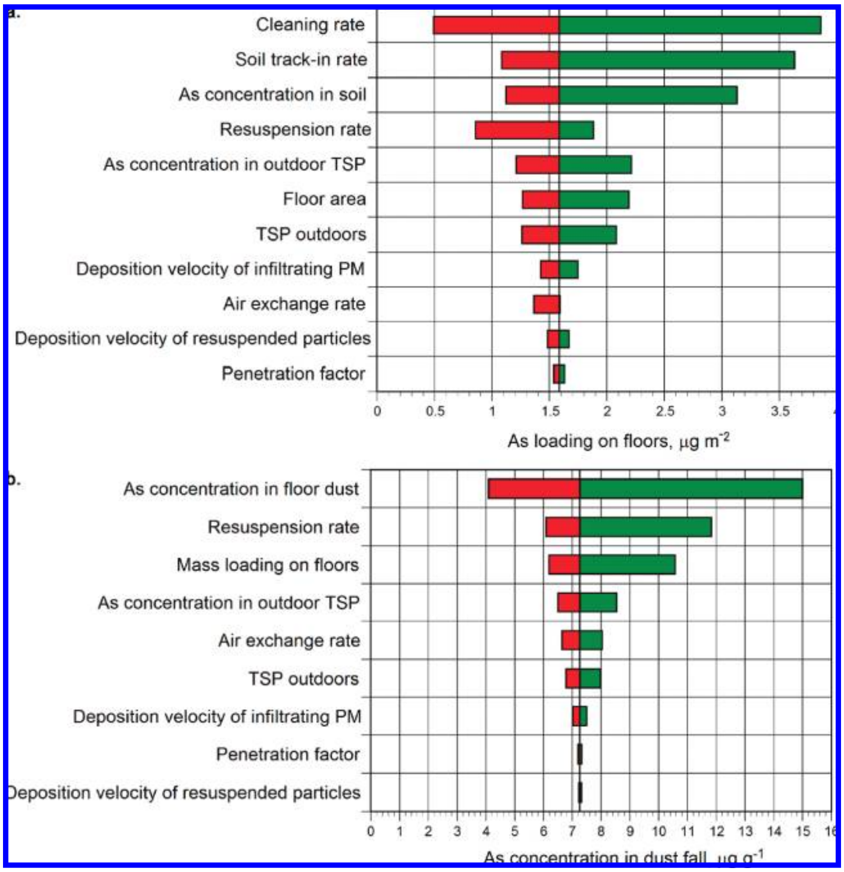

FIGURE 3. Sensitivity analysis of the parameters controlling the (a) predicted level of As loading on floors for Midwest residences and (b) the concentration of As in dust fall. Model responses are determined for low and high values of input parameters, as described in the Supporting Information.

coarse fraction >60 to 150 μm. Particles in the fine fraction preferentially adhere to hands (*40, 41*) and footwear, and importantly, they coincide with the resuspendable dust particles on floor surfaces. Moreover, Edwards et al. (*42*) found that more than 99% of the dust particles deposited on glass slides placed in four New Jersey houses were under 50 μm in size. Deposition velocities calculated for resuspended dust in the NHEXAS Midwest and Arnhem residences (i.e., 175 and 206 m d^{-1}) coincide with these particle sizes as well. Use of the 60 μm cut point demarcating fine particles is also consistent with resuspension experiments conducted with bulk soil samples that demonstrate that soil size fractions <75 μm produce the highest yields of suspended PM less than 10 μm in aerodynamic diameter (*43*).

Although particle adherence to skin and resuspension potential of the coarse fraction of soils/dusts are less than for the fine fraction, the coarse particles may still be important from an exposure standpoint in cases where a contaminant is preferentially enriched on them (*44*). Coarse particles tracked into a residence are redistributed on floor surfaces along with the finer fraction by foot traffic (*45*) and together these two fractions account for over 60% of the dust mass on floors (*46, 47*). The concentrations and particle masses associated with the two size fractions can be used to calculate mass-weighted concentrations representing model parameters (e.g., C_s^s and C_{fl}^s).

One benefit of the dust model is that it establishes the utility of measuring both outdoor TSP and its composition along with soil constituents to simulate indoor dust contamination. Heretofore, relationships between dust contaminants, related human exposures, and outdoor contaminants have been evaluated mainly by empirical methods (*48, 49*). Because air and soil track-in transport pathways can now be analyzed in terms of their individual contributions to dust contamination, risk-management strategies that are pathway specific can also be devised. Additionally, the dust model as currently formulated provides the context for investigating data gaps concerning relationships between human-related factors (e.g., household demographics, cleaning practices and methods, etc.), housing properties (e.g., floor area and coverings, indoor furnishings, etc.) and dust contamination. Further studies are needed as well to evaluate alternative sampling methods for determining the amount of resuspendable dust on various floor surfaces, dust fall rates, and importantly, the duration/frequency of sampling needed to capture time-varying (e.g., seasonal) changes in dust levels and contamination.

Acknowledgments

This work was supported by The University of Arizona, Technology and Research Initiative Fund (TRIF), Water Sustainability Program through the Superfund Basic Research Program (Grant 2 P42 ES04940-11 from the National Institute of Environmental Health Sciences, NIH), and by The University of Arizona, Mel and Enid Zuckerman College of Public Health.

Supporting Information Available

Details on the derivation of the mass-balance equations and the data sources used to quantify model parameters. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES9003735